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## Characterization of polycrystalline materials by X-ray diffraction contrast tomography

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## 1 INTRODUCTION

Synchrotron based X-ray imaging and diffraction techniques offer interesting possibilities for characterizing the grain microstructure in a variety of polycrystalline mono- and multiphase materials. Direct visualization of the three-dimensional grain boundary network or of two-phase (duplex) grain structures by means of absorption and/or phase contrast techniques is possible, but restricted to specific material systems (Ludwig 2009b). However, conventional attenuation or phase contrast imaging techniques do not give access to the crystallographic orientation of the grains and additional diffraction measurements are required. On the other hand, three-dimensional X-ray diffraction methods like Differential Aperture X-ray Microscopy (DAXM, (Larson, 2002) or 3D X-ray Diffraction Microscopy (3DXRD) (Poulsen, 2004) can analyse the 3D grain structure and/or elastic strain tensors of individual grains in polycrystalline materials, but are blind to the microstructural features (inclusions, cracks and porosity) visible in attenuation and/or phase contrast imaging techniques. A recent extension of the 3DXRD methodology, termed X-ray diffraction contrast tomography (DCT) (Ludwig 2009a), combines the principles of 3DXRD and X-ray absorption tomography. With a single scan, DCT can provide simultaneous access to the grain shape, crystallographic orientation, full elastic strain tensor and the local attenuation coefficient distribution in three dimensions. The technique applies to a range of plastically undeformed, polycrystalline mono-phase materials, fulfilling some conditions on grain size and texture. The straightforward combination with in-situ microtomographic observations opens interesting new possibilities for the characterization of microstructure related damage and deformation mechanisms in these materials.

## 2 PRINCIPLE OF X-RAY DIFFRACTION CONTRAST TOMOGRAPHY

X-ray diffraction contrast tomography (DCT) (Ludwig 2008, Johnson 2008, Ludwig 2009a) is a variant of the previously introduced 3DXRD technique enabling simultaneous reconstruction of the 3D grain shapes and orientations in suitable polycrystalline materials. The technique shares a common experimental set-up with conventional synchrotron radiation microtomography. In both cases, the sample is placed on a rotation stage and irradiated by an extended, parallel and monochromatic synchrotron X-ray beam. For the case of polycrystalline materials, each of the grains will pass through Bragg diffraction alignments multiple times during the sample rotation, producing diffracted beams. Beams diffracted at small angles will be captured on the detector system that covers an area substantially bigger than the sample Fig.1. In the absence of orientation and strain gradients inside the grains, the diffracted beams form two-dimensional spots that can be treated as parallel projections of the diffracting grain. The analysis of Friedel pairs of these diffraction spots allows one to determine the crystallographic orientation and 3D shape of the grains in the sample. The processing route can be summarised as follows:

- The diffraction spots are segmented using thresholding techniques and information about the spots is stored in a database (position, intensity, area, etc..)
- From axial symmetry consideration a grain which diffracts for an angular position  $\omega$  diffracts at  $\omega + 180^\circ$  (Friedel pair of diffraction spots corresponding to the  $hkl$  and  $\bar{h}\bar{k}\bar{l}$  reflection of the

same grain). These pairs of diffraction spots (Fig.2a) are detected automatically using a combination of spatial and crystallographic criteria. Once a pair of spots is detected, the diffraction angles describing the geometry of the diffraction event (plane normal, scattering vector) can be calculated (Ludwig 2009a).

- The spotpairs are sorted into sets belonging to the same grain ("indexing"). This is done by checking both spatial and crystallographic consistency criteria. The diffracted beams arising from a grain have to intersect at the grain position, and the angle between scattering vectors has to reflect the crystal symmetry. The set of scattering vectors is used to compute the crystallographic orientation of the grain. For this calculation the Rodrigues space representation (see e.g. (Heinz 1991) of orientation space is used. The resultant Rodrigues vector represents the rotation required to bring the crystal axes into coincidence with the global reference frame.
- In the absence of strong orientation and strain gradients within a grain, the diffraction spots can be considered as projections of the grain from which they arise. They are used to reconstruct the three-dimensional grain shape using algebraic reconstruction techniques (ART) (Gordon, 1977). This algorithm allows the reconstruction of 3D structure from a limited number of projections. Each grain is reconstructed individually (Fig.2b,c). The assembly of all the reconstructed grains produces the 3D grain microstructure of the sample Fig.2d.
- Remaining gaps between grains, as well as overlapping regions in the 3D grain map are removed during a final postprocessing step, in which voxels disputed between several grains are set to zero and grains are dilated until they fill the entire space of the grain map Fig.2e.
- The direct beam images recorded during the scan are used to reconstruct the absorption contrast tomogram of the sample by conventional filtered backprojection reconstruction (Kak, 1988).

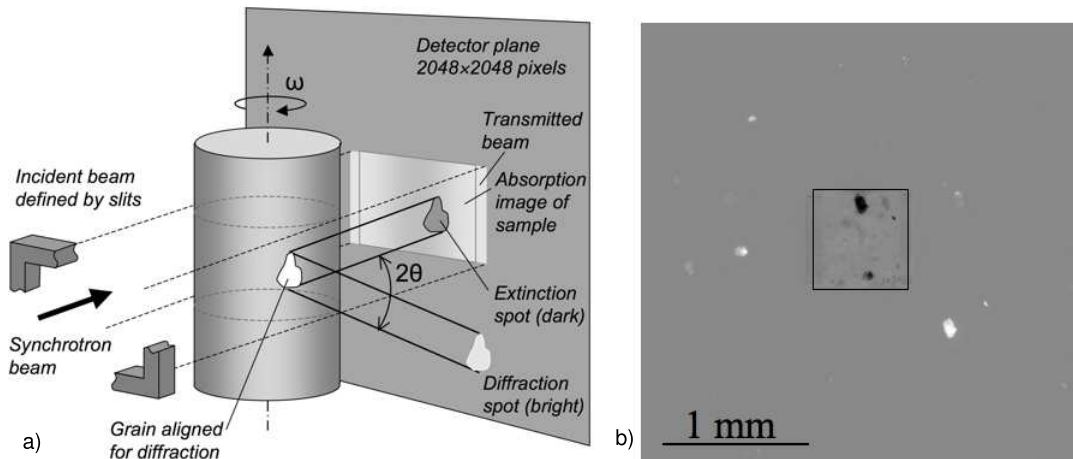


FIG. 1 – Figure 1: Experimental setup for DCT, allowing for simultaneous acquisition of absorption and diffraction information by proximity of sample and detector. b) Example of preprocessed image - remaining contrasts are due to Bragg diffraction of individual grains.

## 2.1 Requirements

The required combination of high spatial resolution and quasi parallel and monochromatic beam illumination of the sample implies that DCT can currently only be performed at synchrotron radiation sources such as the ESRF in Grenoble, France (beam lines ID19 and ID11). Another dedicated instrument is currently under commissioning at the German storage ring PETRA III (Hamburg). Typical acquisition times for a scan on a insertion device beam line like ID19 are of order of 2 hours (7200 projections, 1.4  $\mu\text{m}$  pixel size, 17 keV,  $\frac{\Delta\lambda}{\lambda} = 10^{-4}$ ). Repeated measurement of the time evolution of microstructures subject to annealing or mechanical deformation are feasible but require interruption of the test for the time of the scan acquisition.

Diffraction contrast tomography applies to different classes of mono phase, polycrystalline materials (metals, ceramics, minerals) fulfilling some conditions on grain size, texture and mosaicity. In its current state, the technique works best with materials exhibiting a fully recrystallized microstructure and has only limited capability to resolve sub-grain structures as the ones arising from plastic deformation processes. In order to obtain meaningful shape information, the pixel size  $p$  has to be adjusted to a fraction of the average grain size  $G$  such that  $p \leq \frac{G}{10}$ . The number of grains, and hence the maximum

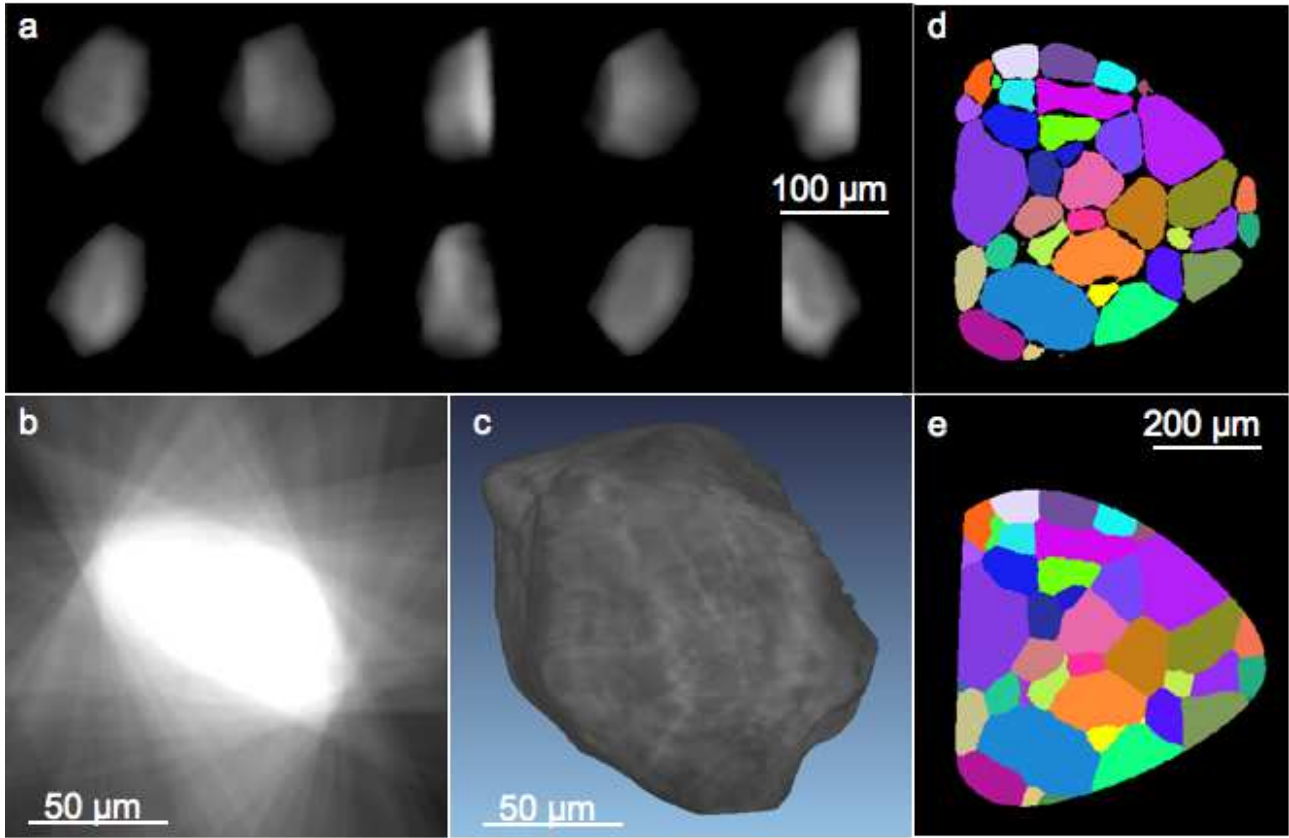


FIG. 2 – a) Set of diffraction spots belonging to one grain. b) approximate shape after first iteration of the iterative 3D ART reconstruction algorithm. c) 3D rendition of the segmented grain volume. d) 2D slice after assembly of the individual grain volumes - overlapping areas are set to zero e) Final reconstruction after dilation postprocessing

sample volume that can be analyzed with a single scan, depend critically on texture and the mosaicity (orientation spread inside grains) of the material. Typical values range from 100 grains for moderately deformed and textured samples, to 2000 and more grains in the case of well re-crystallized microstructures with random texture. The sample diameter is adjusted to meet these requirements.

The need for proximity between detector and sample results in space constraints for the integration of sample environment like furnaces and mechanical test rigs. This problem is most severe when working at high spatial resolution. Miniaturization and design of dedicated test equipment is required for in-situ experiments at this lengthscales. As a parallel beam imaging technique, the ultimate spatial resolution of DCT is limited by currently available detector technology to values of about 1 micrometer. This in turn limits the minimum grain size that can be investigated to about 10  $\mu\text{m}$ .

### 3 APPLICATION EXAMPLES

#### 3.1 Analysis of elastic strain tensors

The Friedel pair acquisition geometry and the fine omega increments used during the scanning procedure (typically  $0.05^\circ$ ) enable us to measure the length and the direction of the diffraction vectors with an accuracy of order of few times  $10^{-4}$  (after correction for systematic errors arising from image distortions and sample drifts). Typically several tens of reflections are available for each grain. By performing a maximum likelihood fit, the six components of the elastic strain tensor can be determined for each of the grains (Reischig, 2008). Fig.3a shows the axial strain  $\epsilon_{33}$ , as measured during a tensile load test in a cylindrical sample made from Ti alloy. The comparison of the measured deformation tensors to the ones predicted from image based crystal plasticity finite element calculations is in progress (Fig.3).

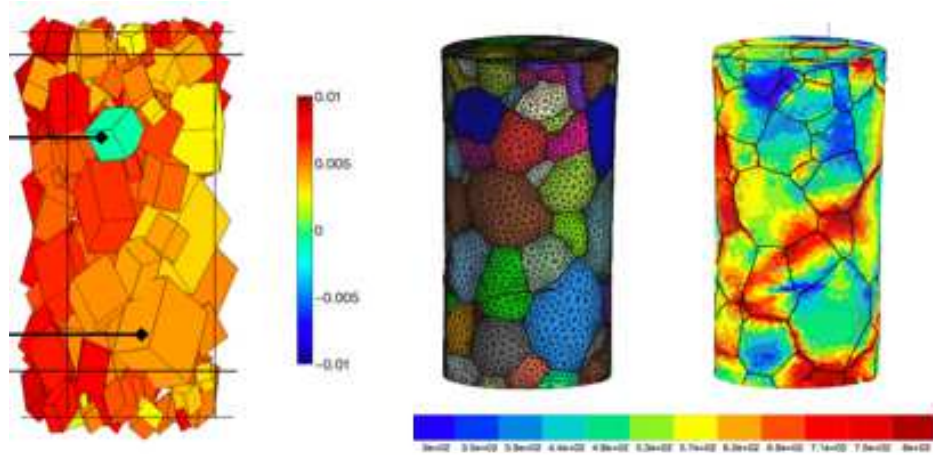


FIG. 3 – a) Axial component of the strain tensor as determined from a DCT scan. b) Image based finite element simulation of the von Mises stress in the same specimen (H. Proudhon, ENSMP)

### 3.2 Orientation Distribution Function

Very much like orientation imaging microscopy (EBSD) does in two-dimensions, DCT gives direct access to the orientation of the grains in 3D sample volumes containing typically  $10^2 - 10^3$  grains. This approach is complementary to established X-ray or neutron diffraction texture measurements, where the orientation distribution function is estimated from a series of pole figure measurements on larger sample volumes containing  $10^4 - 10^5$  grains. Examples of pole figures, calculated from 600 grain orientations and weighted by the corresponding grain volumes are shown in Fig.4. The studied specimen, made from Ti alloy Ti $\beta$ 21s, shows weak signs of rolling texture.

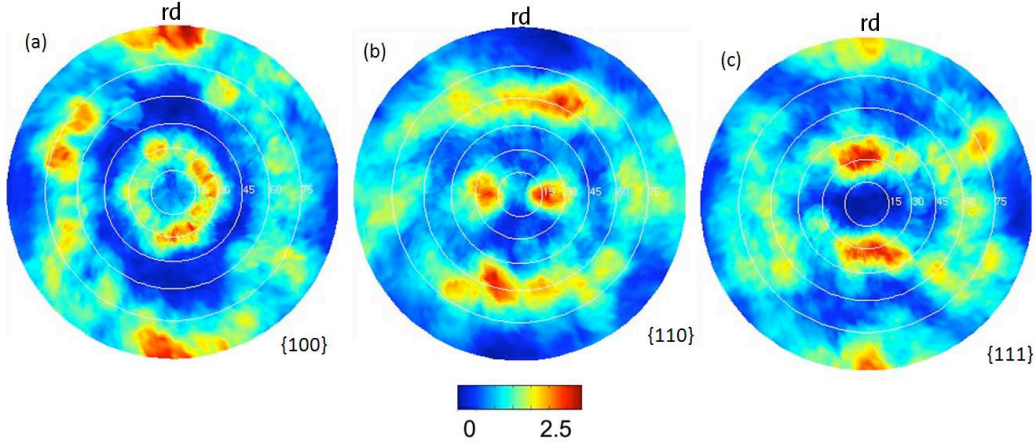


FIG. 4 –  $\{100\}$ ,  $\{110\}$  and  $\{111\}$  pole figures of a weakly textured Ti sample, as calculated from a DCT dataset containing 600 grains

### 3.3 Grain boundary plane distribution functions

The three-dimensional grain volumes produced by DCT give access to a five-parameter description of the grain boundary structure (grain boundary plane orientation together with the crystal misorientation between adjacent grains). The simplest form of analysis involves fitting a plane to each grain boundary, and presenting the distribution of the grain boundary normal vectors in a stereographic projection. The normal directions are defined relative to the crystal axes, and each grain boundary produces two poles in the projection. Another approach, capturing the local curvature of the grain boundaries, consists in meshing the grain boundary network and plotting the orientation densities in the stereographic triangle. This last procedure is illustrated in Fig.5. The surface mesh, color coded with respect to the crystallographic orientation of the grain is shown in subfigure a) and the orientation density plots for the whole grain as well as individual facets of the grain are shown in sub-figures (b-d), respectively. The analysis of grain boundary networks containing a statistically relevant number of facets allows for characterization and subsequent optimization of grain boundary character



distribution via appropriate changes in the thermo-mechanical processing route of the material. This approach, generally known as grain boundary engineering, can for instance be used for optimization of the corrosion resistance of structural steels.

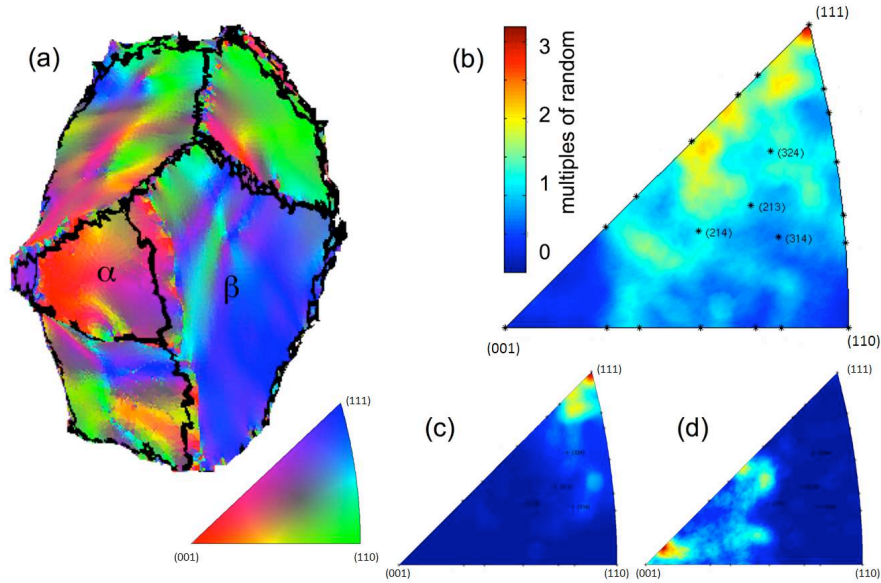


FIG. 5 – a) Grain coloured according to crystal plane of boundary. b) Pole figure of crystal plane of boundary for whole grain. c, d) Pole figures from single grain boundaries marked  $\alpha, \beta$  in (a).

### 3.4 Ongoing research projects

In the course of the last years, the technique has been applied to a variety of materials, addressing questions in different fields of materials science. At the time of writing, part of the projects have given rise to publication, others are in an advanced state of analysis. Here we give a non-exhaustive list of on-going research projects.

- Bulk observation of grain growth in Al and Ti alloys (analysis in progress).
- Observation of inter-granular stress-corrosion cracking in stainless steel (King, 2008).
- Analysis of short fatigue crack propagation in Ti and Mg alloy (analysis in progress)
- Characterization of creep deformation in snow (ANR project 'snow-white', (Rolland, 2010).
- Characterization of grain boundary planes and brittle fracture in Alumina (Marrow, 2009).
- Observation of the onset of plastic deformation in an Al polycrystal (analysis in progress).
- Evolution of elastic strain tensors during tensile loading tests in Ti alloys (Reischig, 2008).

Investigation into (a limited number of) new application areas may be considered on the basis of collaborative research projects and will be subject to the standard ESRF beamtime application procedure.

## 4 PERSPECTIVES

The presence of gradients in orientation and elastic strain inside individual grains lead to a spread of the diffraction spots over an extended angular range, as illustrated in Fig.6 for the case of one reflection from an polycrystalline Alumina sample. The present analysis route only takes average (summed) two-dimensional intensity distributions into account and therefore yields a description of the material in terms of average orientation and elastic strain state for each of the grains. Current work investigates different possibilities to retrieve the information carried by the three-dimensional intensity distribution  $I(x, y, \omega)$  in the diffraction volumes. It can be expected that the next generation of reconstruction algorithms will allow resolving sub-grain orientation and elastic strain distributions at an intermediate lengthscale (resolution element corresponding to a few pixels in the initial grain map). We also investigate into possible combinations of the current full-field imaging approach with scanning micro-diffraction techniques. The combined methodology is expected to provide access to the local variables (orientation and elastic strain) at micrometer spatial resolution. However, the scanning procedure will affect time resolution and its application will have to be limited to a sub-set of grains.

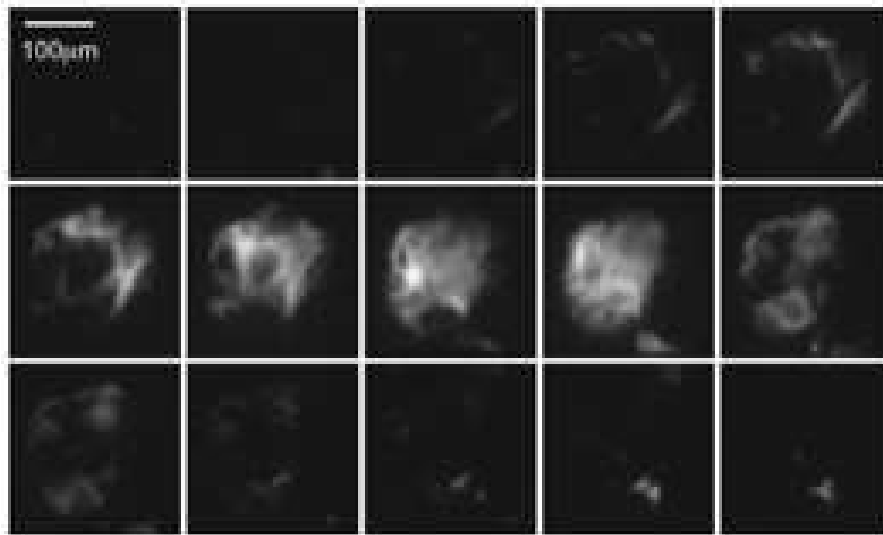


FIG. 6 – Variation in appearance of one diffraction spot at rotation intervals of  $0.05^\circ$ .

## 5 CONCLUSIONS

DCT offers quantitative, comprehensive description of a material's microstructure in terms of 3D grain shape, orientation, elastic strain tensors and local attenuation coefficient distribution. The technique applies to a variety of polycrystalline materials (metals, ceramics, minerals), provided the sample fulfill some conditions (i.e. limited levels of plastic deformation and/or orientation gradients within individual grains). Sharing a common experimental setup, DCT is easily combined with other parallel beam synchrotron imaging techniques like phase contrast micro-tomography. The non-destructive character offers the possibility to observe the response of a polycrystalline material to mechanical deformation and/or annealing processes. The 3D grain maps produced can be used as input for micro-mechanical simulations, taking the grain structure of the material explicitly into account. Direct comparison of model predictions with the experimentally observed material behaviour will become possible.

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